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# Measurement/Evaluation Techniques and Nuclear Data Associated with Fission of $^{239}\text{Pu}$ by Fission Spectrum Neutrons

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# Measurement/Evaluation Techniques and Nuclear Data Associated with Fission of $^{239}\text{Pu}$ by Fission Spectrum Neutrons

Joint LANL/LLNL Fission Product Evaluation Review Panel

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## 1 Executive Summary

This Panel was chartered to review and assess new evaluations of work on fission product data, as well as the evaluation process used by the two U.S. nuclear weapons physics laboratories. The work focuses on fission product yields resulting from fission spectrum neutrons incident on plutonium, and includes data from measurements that had not been previously published as well as new or revised fission product cumulative yield data, and related quantities such as Q values and R values.<sup>1</sup> This report documents the Panel's assessment of the work presented by Los Alamos National Laboratory (LANL) and Lawrence Livermore National Laboratory (LLNL). Based on the work presented we have seven key observations:

1. Experiments conducted in the 1970s at LANL, some of which were performed in association with a larger, NIST-led, program, have recently been documented.<sup>2</sup> A preliminary assessment of this work, which will be referred to in this document as ILRR-LANL, shows it to be technically sound.
2. LLNL has done a thorough, unbiased review and evaluation of the available literature and is in the process of incorporating the previously unavailable LANL data into its evaluation of key fission product yields. The results of the LLNL effort, which includes a preliminary evaluation of the ILRR-LANL data, have been documented.<sup>3</sup>
3. LANL has also conducted an evaluation of fission product yields for fission spectrum neutrons on plutonium including a meta-analysis of benchmark data as part of a planned upgrade to the ENDF/B compilation. We found that the approach of using meta-analysis provides valuable additional insight for evaluating the sparse data sets involved in this assessment.

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<sup>1</sup>See Glossary for definitions of terms.

<sup>2</sup>Mac Innes, M. R. et al., "Fission Product Data Measured at Los Alamos for Fission Spectrum and Thermal Neutrons on  $^{239}\text{Pu}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$  (DRAFT)", LA-UR 09-06679, Nov 2009.

<sup>3</sup>Henderson, R. A. (editor) et al., "Fission Chain Yield Evaluation Report", LLNL-TR-418425-DRAFT, Nov. 2009.

4. Both laboratories have provided convincing evidence for energy dependence in the fission product yield of  $^{147}\text{Nd}$  produced from the bombardment of  $^{239}\text{Pu}$  with fission spectrum neutrons over an incident neutron energy range of 0.2 to 1.9 MeV.
5. Consistent, complete, and explicit treatment of both systematic and statistical uncertainties, including correlations, are critical to the assessment of both the experimental measurements (due to variations between experimental techniques, irradiation conditions, calibration procedures, etc.), and the evaluation of those experiments to extract fundamental nuclear data. A clear example of the importance of uncertainty analysis is in the justification for energy-dependent  $^{147}\text{Nd}$  fission product yield, where the magnitude of the effect is comparable to the uncertainties of the individual fission product yield measurements. Both LANL and LLNL are committed to the inclusion of full uncertainty analysis in their evaluations.
6. The Panel reviewed in detail two methods for determining/evaluating fission product yields from which fission assessments can be made: the K factor method and high-resolution gamma spectroscopy (both described more fully in Sections 3 and 4). The panel concluded that fission product yields, and thus fission assessments, derived using either approach are equally valid, provided that the data were obtained from well understood, direct fission measurements and that the key underlying calibrations and/or data are valid for each technique.
7. The Panel found the process of peer review of the two complementary but independent methods to be an extremely useful exercise. Although work is still ongoing and the numbers presented to the Panel may change slightly, both groups are now in much better agreement on not just one, but four key fission product yields. The groups also have a better appreciation of the strengths and weaknesses of each other's methods.

Draft fission product yield evaluations ( $Y_j^{239\text{Pu},fs}$ ;  $j = ^{95}\text{Zr}, ^{99}\text{Mo}, ^{144}\text{Ce}$ , and  $^{147}\text{Nd}$ ) for “fission spectrum” neutrons on  $^{239}\text{Pu}$  reported by both laboratories are now in good agreement (See Table 1 in Appendix A). The laboratories are also in closer agreement on the inferred  $Q_{^{99}\text{Mo}}^{239\text{Pu},fs}$  (See Table 2 in Appendix A). Modest further work may reduce differences even further.

The structure of this document is as follows: Following a brief introduction, Section 3 gives some background information related to each laboratory's fission measurement technique. Sections 4–7 provide additional detail related to points 1–5 in the Executive Summary. A Recommendations and Conclusion section summarizes the material presented, and two Appendices and a Glossary are included for reference.

## 2 Introduction

We compliment the teams from both laboratories on their perseverance in finding and analyzing data from experiments that were done, in many cases, decades ago. This has included searching through old laboratory notebooks, interviewing people long retired and delving into reports that were never published in the open literature<sup>4</sup> from laboratories across the world. This level

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<sup>4</sup>In many cases work was documented via institutional/internal reports.

of scientific archeology was warranted both because of the importance of these values to the laboratories and because of the paucity of data.

This research provides compelling evidence for changes away from the values used by both laboratories prior to the 1990's in analyzing key radiochemical data used for the determination of the number of fissions in a sample. Both the new experimental data available for today's evaluations as well as the evidence of energy dependence in  $^{147}\text{Nd}$  support these conclusions.

We *strongly* encourage the publication of these results in the peer-reviewed scientific research literature. The fact that not all of the experimental details, as well as some of the results, were not readily available to both laboratories or the broader scientific community has not served us well, and must not continue.

The laboratories are now very close to agreement on critical values for key fission products. Agreement on these values would provide an "island of stability" for both laboratories. We encourage both teams to use the ongoing ENDF process as a mechanism by which their data and evaluations may be integrated into the larger nuclear data community.

We recognize and applaud the significant efforts both laboratories have made in addressing and treating experimental uncertainties. Because of the very complicated nature of this issue the Panel urges both groups to re-examine their treatment of these uncertainties, and to make sure their treatment is a key part of the papers submitted for publication. Further, we urge that once this re-examination is completed the groups continue their technical exchange to reconcile and document their results.

### 3 Background

In the early days of fission analysis, there were two significant obstacles to quantitative measurements: 1) stable radiation detectors and 2) lack of detailed knowledge regarding many fundamental nuclear decay phenomena. It was known, however, that binary fission was the primary mode of neutron-induced nuclear scission, that the fission process resulted in the production of a bimodal distribution of nuclides, centered approximately (for  $^{235}\text{U}$ ) around masses 96 and 140, and that fission products decayed via multiple pathways involving emission of  $\beta^-$  particles and  $\gamma$  rays. This information was employed to develop a method for determining the number of fissions in any given sample that was robust with respect to the two issues noted above.

It was recognized that if a "standard" fission product source could be identified, both concerns could be mitigated using ratio measurements.  $^{235}\text{U}$  was available, as was a research reactor with a region where neutrons were moderated to the thermal environment of the reactor. It was known that  $^{99}\text{Mo}$  was produced with significant yield and decayed via  $\beta^-$  emission. Further, highly selective chemical separations for the isolation of Mo from all other elements were known. Finally, the short half-life of  $^{99}\text{Mo}$  resulted in significant count rates in proportional counters even for samples containing small amounts of fission. Thus, determination of the  $^{99}\text{Mo}$   $\beta^-$  emission count rate resulting from irradiation of  $^{235}\text{U}$  in a thermal neutron field was a convenient "standard reference material" for quantitative fission measurements.

An independent measurement of sample fissions, accomplished using a fission chamber co-located in the same neutron field as the  $^{235}\text{U}$  sample, provided the "absolute" scale for the method, and the K factor is the numerical statement of that scale (fissions/cpm). In the time

when the service life of radiation detectors was short (perhaps only days) it was necessary to determine the K factor for almost every measurement. As detector technology improved, so did equipment stability and longevity.

Count rates on different detectors could be compared by counting the same sample on each. Relative count rates (r values) for different fission product nuclides (chemically separated and counted independently) from the same sample could be tabulated, providing an ability to assess relative fission product content. For neutron fields other than thermal, and for targets other than  $^{235}\text{U}$ , K factors could also be determined. A mathematical framework was developed to relate r values and K factors such that for an arbitrary sample, the total fission content could be inferred. These relationships all hinge, ultimately, on the  $^{235}\text{U}$  thermal K factor; hence its importance in the establishment of the laboratory's calibration.

The “historic” LANL fission scale was codified by Browne in 1956. In the mid-1970s, discrepancies between historic LANL values and measurements associated with the ILRR collaboration (see Section 4) led to the discovery of an error in LANL historic thermal K factors that has been attributed to uncompensated self-attenuation in the target samples. A new set of K factors, consistent with the ILRR results, was first presented by LANL in 1977 but not adopted until 1997. These K factors define the “modern” LANL fission scale.

When the Lawrence Livermore National Laboratory (LLNL) opened in 1952, the only established method known in the US for generating fissions in a sample from the measurement of fissions products was the K factor method employed at Los Alamos. In the late 1960s and early 1970s, with the development and continued sophistication of gamma spectroscopy, LLNL chose to depart from the LANL K factor method and move to a method that utilized the growing availability of nuclear data and the improvements in germanium detectors. The gamma spectroscopy method requires identification and quantification of the specific nuclear data and detector characteristics that relate the measured photons from decay of a fission product directly to the number of fissions in a sample. These components include nuclear data (e.g., half-lives, branching ratios, etc.) and counter response (e.g., efficiency, background, etc.).

To obtain the required nuclear data, LLNL adopted a method of evaluating nuclear data, both published and unpublished, from global sources and conducting reevaluations as new data became available. The counter response for the gamma spectrometers were obtained by extensive and meticulous calibration procedures using known standards. Because the gamma spectroscopy method uses evaluated nuclear data and calibrated detector efficiencies to convert count rates to absolute numbers of atoms of a given fission product in a sample, a data reduction code, GAMANAL, was developed at LLNL. Over the years the GAMANAL code has been adopted by many research groups and continues to be widely used around the world, most notably by the IAEA in the area of nuclear safeguards. Relating the number of fission product atoms to number of fissions using the gamma spectroscopy method requires additional data, namely fission product chain yields.

Both methods have strengths and weaknesses but since they are complementary yet independent, they provide additional confidence when both methods converge on the same answer when applied to the same sample. The strength of the K Factor method is that a self-consistent data set can be constructed but it hinges on getting the K factor correct for  $^{99}\text{Mo}$  ( $^{235}\text{U}$  thermal). Any error in this value is propagated through all of the other fission products since they are measured relative to  $^{99}\text{Mo}$ . The K factor method also requires careful attention to and documentation of internal laboratory calibration. The major limitations of the gamma spectroscopy

method include the requirement to have certain pieces of nuclear data known with reasonable accuracy and precision (e.g., branching ratios and fission chain yields) and counter responses (e.g., background and efficiencies). On the other hand, the strengths of the gamma spectroscopy system is that in many instances, labor-intensive chemical separations can be eliminated, many nuclides can be measured from a single sample, and data can be easily reanalyzed when new nuclear data are introduced, such as branching ratios or interfering gamma lines.

## 4 Assessment of LANL Experimental Methods and Documentation

We conclude that the data from the experiments performed by LANL are well documented in LA-UR 09-06679, are sound, and should be included in the over-all assessment of fission chain yields.

### 4.1 Beta Counting – K/Q/R methods

From the early days of the laboratory, LANL has relied primarily on beta activity counting of chemically separated fission products (FPs) for the determination of total fissions in critical assembly tests and other samples. Although fission product yields of specific FPs can be derived from these results, fission product yield data and gamma branching ratio data were not needed for the determination of total fissions in the LANL beta analysis method. Fissions were determined by K factors that related FP activities to fissions, by ratios of K factors called Q values, and by beta activity ratios called r (“little” r) and R (“big” R) values (see Glossary). The long term reliance on these beta analysis methods was continued not just for the sake of traceability to historic experiments, but because several systematic uncertainties in nuclear data and detector efficiency data could be eliminated. As modern gamma-ray spectroscopy technology developed, the beta counting approaches were supplemented by gamma-ray spectroscopy to make up for some of the limitations of the beta analysis method, in particular the detection of trace levels of activities of other FPs that might not be distinguishable by half-life observations alone. Absolute determinations of fissions in every neutron spectrum of interest and for every fissionable isotope of interest did not have to be done, because the detection efficiency and half-life for a particular FP are independent of the neutron spectrum inducing the activity. The beta activity analysis method was tested in frequently repeated experiments and found to give reliable and consistent results over periods of several decades. The maintenance of the experimental apparatus and the documentation of the experiments has been excellent, with one notable exception: the  $^{239}\text{Pu}$  macrofoil target diameter for thermal neutron field irradiations was not preserved, resulting in an uncertainty associated with the mass thickness of those deposits. However, Q values derived from those measurements have been superseded by the experiments documented in LA-UR 09-06679, and described further below.

While these methods are different than those based on  $\gamma$  spectra that developed as Ge(Li) detectors became available, they appear to have comparable validity in determining the number of fissions that occurred. Furthermore, comparisons of the chemically separated  $\beta^-$  activities with  $\gamma$ -ray spectroscopy measurements at LANL have alleviated concerns about possible contaminants in the chemical separations.

The LANL radioanalytical measurement system is calibrated on the basis of K/Q/R measurements, and the LANL “historic” scale was established as discussed in Section 3. A major revision of some of the key Q values was found to be necessary as a result of measurements performed in the mid-1970s. The NIST<sup>5</sup>-led ILRR<sup>6</sup> collaboration undertook very careful fission product yield determinations in several “fission spectrum” neutron fields, including the LANL Big-10 critical assembly. The ILRR program used  $\gamma$  spectroscopy analysis at multiple laboratories for their fission product assessments; these results will be referred to as ILRR-NIST. LANL radiochemistry participated in these experiments, but applied their standard methodologies (i.e. chemical separation followed by beta counting) to the measurement of FP activities, and those results are denoted as ILRR-LANL. The revision of the LANL Q values was stimulated by the discrepancies between the ILRR-LANL results and “historic” LANL Q values.<sup>7</sup> The ILRR-LANL tests led to the establishment of “modern” LANL Q values<sup>8</sup> with careful attention to self-shielding in the thermal neutron irradiations. The discrepancies between the “historic” and “modern” values were ascribed largely to previous errors in corrections for self-shielding. The “modern” Q values stand on the 1970s experiments and are not directly dependent on the explanation of the previously discrepant results. Fission product yields inferred from the “modern” LANL Q values are consistent with the ILRR-NIST published results for those FP nuclides that were measured by both groups.

The documentation of LANL’s basis for fission determinations has included examination of extensive LANL documentation, examination of the voluminous ILRR documentation, and interviews with retired staff, semi-retired staff, and retired collaborators from the ILRR experiments. Improvements remain to be made in reporting full uncertainty budgets for the K/Q/R method, particularly in the propagation of uncertainties in the derived fission product yield results, with attention to major correlations and separate reporting of statistical and systematic uncertainty components, as well as the combined uncertainties.

## 4.2 Accounting for <sup>240</sup>Pu Content in LANL R values

The LANL 1950s R value measurements at Flattop and other critical facilities were done with Pu macrofoils containing  $\approx 6$  atom% <sup>240</sup>Pu. LANL chemists demonstrated that the influence of spontaneous fission from this isotope on the fission product counting was negligible. However, the potential bias of neutron-induced fission of <sup>240</sup>Pu should be taken into account for comparisons with pure <sup>239</sup>Pu measurements. The magnitude of the effect is, not surprisingly, dependent on the details of the neutron field (noting that the <sup>240</sup>Pu (n,f) cross section has a threshold of  $\approx 0.7$  MeV). Due to the one mass unit excess some fission product yields can be significantly different. Experimental measurements of <sup>240</sup>Pu FP yields in a fission spectrum neutron field were published in the late 1970s.<sup>9</sup> The fission product yields reported therein are larger by about 10% than the LANL values for <sup>239</sup>Pu in a fission spectrum in the case of <sup>147</sup>Nd and <sup>144</sup>Ce, whereas in the case of <sup>95</sup>Zr, the yield is smaller by about 10%. The yield for <sup>99</sup>Mo is essentially unchanged between <sup>239</sup>Pu and <sup>240</sup>Pu. Appendix B details the process for estimating the impact

<sup>5</sup>At that time NBS (National Bureau of Standards).

<sup>6</sup>Interlaboratory LMFBR (Liquid Metal Fast Breeder Reactor) Reaction Rate

<sup>7</sup>“Historic” LANL  $Q_{99\text{Mo}}^{239\text{Pu},fs} = 0.966$

<sup>8</sup>“Modern” LANL  $Q_{99\text{Mo}}^{239\text{Pu},fs} = 1.015$  (see also Table 2, 1st column)

<sup>9</sup>Myers et al., “Fast-Neutron Fission of <sup>240</sup>Pu”, *Phy. Rev. C* **18**(4), 1700 (1978).

of  $^{240}\text{Pu}$  content on both fission product yields and R values. Although the magnitude of the corrections are small (0.3–0.6%), the treatment of this bias was not handled consistently among laboratories, so care must be taken when making comparisons of plutonium fission product data.

## 5 LLNL Evaluation of Global Experimental Literature

The key conclusions from the LLNL evaluation include conclusive evidence to change the fission chain yield data in use at LLNL. The direction and magnitude of the changes put the two laboratories in significantly better agreement for the important peak yield fission products ( $^{95}\text{Zr}$ ,  $^{99}\text{Mo}$ ,  $^{144}\text{Ce}$ ,  $^{147}\text{Nd}$ ) than prior to the evaluation, generally  $\ll 1\%$  (see Table 1); the  $^{99}\text{Mo}$  values are most disparate at  $\approx 2\%$  in these draft reports and further work may improve the agreement. As discussed previously, both laboratories need to better address uncertainties in their final reports, but the agreement between the two for most isotopes is likely to be within uncertainties.

For  $^{95}\text{Zr}$ ,  $^{99}\text{Mo}$  and  $^{144}\text{Ce}$ , the result from the LLNL draft report is a  $\approx 5\%$ ,  $\approx 3\%$  and  $\approx 8\%$  increase from 4.52 to 4.77, 5.94 to 6.12 and 3.34 to 3.69, respectively. For  $^{147}\text{Nd}$ , while the LLNL value (1.97) is consistent with fast reactor data, the presence of an energy dependence will likely effect a change in the use of the data. The energy dependence is approximately a 4% change in the fission chain yield value per MeV of incident neutron energy.

Similar to the 147 mass chain, there may be a relatively weak energy dependence associated with the 144 mass chain. Uncertainties on the available data were too high to definitively determine any effect; while a dependence was observed, the uncertainty on that slope was equal to the measurement uncertainty. For the  $^{95}\text{Zr}$  and  $^{99}\text{Mo}$  data sets, there was no statistically significant energy dependence.

We compliment LLNL on a thorough, unbiased review of the available literature. The inclusion of the draft version of the ILRR-LANL data was particularly welcome. A revised LLNL Review Document should be issued upon receipt/review of the “final” version of LA-UR 09-06679. We recommend that the final version of the LLNL Review Document contain the following features:

- Clear statements regarding where, in what way, and how results were adjusted between those in the literature and those presented in the Review Document, including where gamma intensity values have been updated.
- Clear statements indicating where data were insufficient to allow full verification and validation of the literature values, including a statement on the possible impact of the lack of data on the quoted results.

Special consideration should be given to:

- The impact of systematic uncertainties on the results.
- Identifying correlations within and between the reviewed data sets.

A decision regarding how to best employ the [Laurec 81]<sup>10</sup> data should be addressed. Appropriate use of these data strengthens the evaluation process. More specifically, these points need to be addressed:

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<sup>10</sup>Laurec, J. et al., “Détermination des Rendements de Fissions Induites par un Spectre de Neutrons de Fission dans le  $^{239}\text{Pu}$  et l’  $^{235}\text{U}$  pour les chaînes 95, 144 et 147”, CEA report, CEA/DE/RCP/DO-00058, Jan 1981.

- Update of the gamma intensity values using the most recent published nuclear data.
- An accounting of the systematic uncertainties applied to these new gamma intensity values (the uncertainty values quoted in [Laurec 81] do not include the systematic uncertainty due to gamma intensity. In the case of the  $^{147}\text{Nd}$  fission spectrum yield of  $^{239}\text{Pu}$ , the quoted uncertainty is 4%, to which a  $\approx 5\%$  uncertainty due to gamma intensity must be added in quadrature resulting in a  $\approx 7\%$  compound uncertainty on the  $^{147}\text{Nd}$ . However, when considering ratios (R, Q, and the ratio to  $^{140}\text{Ba}$ ) the original quoted uncertainties must be used (not the compound ones including the separate uncertainty of  $I_\gamma$ ) because the same systematic uncertainty will be present for both the numerator and the denominator, and thus cancel out.

Finally, as this review document will likely become an important reference for the evaluation community, we recommend that the basic experimental data used for LLNL's assessment be documented in a form that includes values, uncertainties, and (if possible) covariances.

## 6 Meta-Analysis for Improving the Statistical Population Available for Evaluation

Part of the LANL evaluation involved use of a “meta analysis” to verify and refine the evaluation of the Q-value for  $^{99}\text{Mo}$  resulting from irradiation of Pu with “fission spectrum” neutrons ( $Q_{^{99}\text{Mo}}^{^{239}\text{Pu},fs}$ ).<sup>11</sup> This technique allows incorporation of a much wider range of experimental data, which is especially important for  $^{99}\text{Mo}$  because of the small amount of direct experimental data available. Specifically, LANL was able to incorporate R values from  $^{140}\text{Ba}$ ,  $^{95}\text{Zr}$ ,  $^{97}\text{Zr}$ ,  $^{137}\text{Cs}$ ,  $^{141}\text{Ce}$  and  $^{143}\text{Ce}$  from experiments by Maeck, Lisman, Laurec, ILRR-LANL, and ILRR-NIST in determining  $Q_{^{99}\text{Mo}}^{^{239}\text{Pu},fs}$ . Full consideration of energy-dependencies was included, largely by concentrating on fission products assumed to have minimal energy dependence. The impact of incident-energy fission product yield dependence was assessed and essentially no change in the results was found. Additionally, a full account of statistical and systematic uncertainties was included in the analysis.

The results of this analysis were shown to be completely consistent with the direct measurements and were subsequently incorporated into the final analysis. The Panel endorses this approach to widening the applicable database for the  $Q_{^{99}\text{Mo}}^{^{239}\text{Pu},fs}$  analysis. It is recommended that details of the analysis with uncertainties and correlations be fully documented and published.

Use of the Laurec data in this evaluation reveals some intriguing features. The meta-analysis of the data in terms of  $Q_{^{99}\text{Mo}}^{^{239}\text{Pu},fs}$  values seems to indicate that there is a systematic disagreement with the other  $Q_{^{99}\text{Mo}}^{^{239}\text{Pu},fs}$  values (both measured and derived) used in the evaluations. On the other hand, R values constructed for Laurec as well as ratios of the  $^{140}\text{Ba}$  (both of which can be considered as a way of getting rid of the absolute counting of fissions) seem to fall in line with the bulk of the data considered in the evaluations. We find that the Laurec R values should be included in the analysis.

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<sup>11</sup>Chadwick, M. B. et al., “Evaluation of Fission Product Yields from Fission Spectrum n+ $^{239}\text{Pu}$  Including a Meta Analysis of Benchmark Data: ENDF/B-VII.1 Upgrade,” LA-UR-09-04234, November 2009.

## 7 Assessment of Evidence Related to Energy-Dependent Fission Chain Yield in $^{147}\text{Nd}$

Both LANL and LLNL have done statistical analyses of the energy dependence of the  $^{147}\text{Nd}$  fission product yield for  $^{239}\text{Pu}$  over the neutron energy range from 0.2 MeV to 1.9 MeV. LANL examined FP yields and R values and the ratios of  $^{147}\text{Nd}$  yields to  $^{99}\text{Mo}$ , and found energy dependence. LLNL extended the studies by examining activity ratios with  $^{140}\text{Ba}$  data, thus increasing the relevant data and found a similar result.

These analyses provide convincing evidence that the  $^{147}\text{Nd}$  fission yield changes with a small positive slope over the energy range studied. The two analyses are in substantial agreement over the magnitude of the slope. They also showed a very small probability that there was no energy dependence - they resoundingly reject the null hypothesis that the slope is zero. This energy dependence is established only for the limited energy range studied, and should not be applied outside the range. A similar study on the zirconium data showed no discernible variation for the  $^{95}\text{Zr}$  fission product yield.

The experimental data were obtained from both critical assemblies and fast reactor experiments, none of which are mono-energetic neutron sources. Thus each data point represents an average over an appreciable energy range. The analysis used by both laboratories involved semi-empirically fitting a linear model through the evaluated data; thus, no structure in the energy dependence could be inferred. The results could be looked at as a measure of the first moment of the real structure, or as the first term in a Taylor series expansion.

In addition, we saw a presentation by Stan Prussin, representing a LLNL-commissioned group of independent reviewers. Their study was based on a subset of the available data over a more restricted energy range, focusing on measurements of relatively high precision obtained for several isotopes. The results presented were consistent with, though not altogether independent of, both labs' findings. They indicate self-consistency regarding energy dependence of the mass 147 yield. Because we had less information about this work than that presented by the laboratories, we were unable to evaluate it as thoroughly. We encourage this group to seek publication of their findings.

## 8 Conclusions and Recommendations

The Joint LANL/LLNL Fission Product Evaluation Review Panel has received and considered both written and oral input from the laboratories related to key nuclear physics measurements and evaluations of selected fission product data. The Panel believes that the results of these significant efforts, specifically the documentation of the experimental work at LANL in the 1970s and the evaluation by LLNL of the relevant available literature, form the basis for revising  $Q_{^{99}\text{Mo}}^{^{239}\text{Pu},fs}$ , which impacts LANL data evaluations.<sup>12</sup> Such a change effectively revises fission product chain yields, impacting data evaluations based on analyses of  $\gamma$  spectra (performed at both LLNL and LANL).

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<sup>12</sup>The  $Q_{^{99}\text{Mo}}^{^{239}\text{Pu},fs}$  given in the first column of Table 2 has been in use at LANL since the mid-1990s.

To summarize the Panel’s conclusions and recommendations:

1. **Recognition of both laboratories’ work** - We wish to recognize and commend both Laboratories for their extensive efforts to address long-standing issues related to nuclear data associated with fission. We urge both laboratories to complete their analyses as soon as possible, and to submit their work to an appropriate peer-reviewed journal for publication.
2. **ILRR-LANL** - We conclude that the data from the experiments performed by LANL are well documented in LA-UR 09-06679, are sound, and should be included in the over-all assessment of fission chain yields.
3. **LLNL Literature Review** - We conclude that LLNL has performed a thorough and unbiased review of the relevant literature data for the fission chain yields ( $Y_j^{i,e}$ ) for selected fission products resulting from neutron induced fission of  $^{239}\text{Pu}$  with “fission spectrum” neutrons.
4. **Meta-Analysis** - The meta-analysis approach allowed both Laboratories to expand the set of data available for the evaluation of fission product chain yields for  $^{239}\text{Pu}$  in a fission spectrum neutron field. Although each Laboratory took different approaches in both the evaluation of the data and the associated uncertainties, their conclusions were consistent with each other. However, the treatment of correlated uncertainties merits further consideration by both groups. We urge both LANL and LLNL to engage and take advantage of experts in statistical analysis in addressing this issue.
5. **Energy Dependence** - We found convincing experimental evidence for energy dependence in the production of  $^{147}\text{Nd}$  from the irradiation of  $^{239}\text{Pu}$  with fission spectrum neutrons. Theoretical work is needed to provide fundamental understanding of this observation, and to suggest a functional form for fitting empirical data. Knowing the functional form of this dependence would be important for reducing the uncertainty associated with the application of this fission product indicator. Additional experiments, especially in the range of 2-6 MeV, would be helpful in addressing this issue and constraining theoretical models.
6. The Panel reviewed in detail two methods for determining/evaluating fission product yields from which fission assessments can be made:
  - (a) The first is called the K factor method. The K factor is a constant that relates the measured decay rate of a fission product directly to the number of fissions in a sample. Historically, the K factor method relied on beta activity counting of chemically separated samples of fission products (FPs) using a  $\beta^-$  counter that was well calibrated in fission chamber experiments;  $^{99}\text{Mo}$  produced from the thermal neutron irradiation of  $^{235}\text{U}$  was taken to be the standard. Other fission products were then determined relative to  $^{99}\text{Mo}$  through ratio measurements (r values) of activities (count rates) from the same source material. The K factor method requires that K factors be measured for each fissioning system as well as in each neutron spectrum inducing fission. However, once the K factors are known, ratios of K factors (Q values) and

ratios of ratios of fission product activities relative to the standard (R values) are used to interrelate fission products. Although fission product yields were not determined directly by this method, fission product yields could be derived and compared to other fission product yield measurements by assuming/adopting a value for the fission product yield of the standard. The  $^{235}\text{U}$  thermal fission product yield for  $^{99}\text{Mo}$  from the ENDF/B-VI compilation is a quantity that has been reported by many investigators and is considered to be well characterized.

- (b) The other method relied on measuring fission product yields directly by gamma spectroscopy using high resolution germanium detectors, both with and without chemical separation, and converting the measured activity (count rate) to atoms using detector efficiencies and decay scheme information such as branching ratios. Again, an independent measurement (e.g. using a fission chamber) is needed to assess total fissions. By taking ratios of fission product yields, Q values can be derived and compared to those from the K factor method.

The Panel concluded that fission product yields and thus, fission assessments, derived using either calibrated K factors from counting fission products in chemically purified samples or directly from gamma spectroscopy of samples (with or without chemical separation) are equally valid, provided that the data were obtained from well understood, direct fission measurements and:

- (a) For the K factor method, the laboratory calibration is maintained;
  - (b) For the gamma spectroscopy method, the detector efficiencies, half-lives, and branching ratios are well known.
7. The Panel found the process of peer review of the two complementary but independent methods to be an extremely useful exercise. Although work is still ongoing and the numbers presented to the Panel may change slightly, both groups are now in much better agreement on not just one, but four key fission product yields. The groups also have a better appreciation of the strengths and weaknesses of each other's methods.

## A Nuclear Data Associated with Fission

The Panel believes it is important to capture a “snapshot” of the current state of several key pieces of nuclear data as measured and/or evaluated by the two laboratories. This will allow for more facile comparisons in the future as evaluations are updated. Table 1 presents, from documentation provided to the Panel by the laboratories, comparisons of evaluated fission product yields for  $^{95}\text{Zr}$ ,  $^{99}\text{Mo}$ ,  $^{144}\text{Ce}$ , and  $^{147}\text{Nd}$ , the latter being reported at a “mean energy of neutron causing fission” ( $\langle E_n \rangle$ ) of 1.5 MeV. It should be noted that these numbers are from documents that are presently in DRAFT form; archival documents should be consulted for authoritative values.

Table 1: Comparison of selected evaluated fission product yields ( $Y_j^{239\text{Pu},fs}$ ) produced by bombardment of  $^{239}\text{Pu}$  with “fission spectrum” neutrons. The subscripted final digit in some entries indicates an additional, non-significant, figure that is carried to avoid truncation errors if the values are used in further calculations.

Nuclide ( $j$ )	Evaluated Fission Product Yield (%)		LANL/LLNL
	LANL <sup>13</sup>	LLNL <sup>14</sup>	
$^{95}\text{Zr}$	$4.76 \pm 0.03_3$	$4.77 \pm 0.02_8$	$0.998 \pm 0.009$
$^{99}\text{Mo}$	$6.23 \pm 0.06$	$6.12 \pm 0.08$	$1.018 \pm 0.017$
$^{144}\text{Ce}$	$3.69 \pm 0.02_6$	$3.69 \pm 0.01_8$	$1.000 \pm 0.008$
$^{147}\text{Nd}^{15}$	$2.10 \pm 0.02_9^{16}$	$2.07 \pm 0.03_9^{17}$	$1.014 \pm 0.024$

The experimentally measured calibration basis at LANL is the Q value for  $^{99}\text{Mo}$ , *which is not dependent upon knowledge of fission chain yields*. This value is tabulated for targets of

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<sup>13</sup>LA-UR 09-04234, Table 13

<sup>14</sup>LLNL-PRES-419362, Slide 84

<sup>15</sup> $\langle E_n \rangle = 1.5$  MeV

<sup>16</sup>The LANL energy dependence relationship, from p. 25 of LA-UR 09-04234, is:

$$Y_{147\text{Nd}}^{239\text{Pu},fs} = [(0.097 \pm 0.027) \cdot \langle E_n \rangle] + (1.957 \pm 0.014)$$

Due to correlations within the data used in the development of this model, the uncertainty reported in Table 1 is smaller than what is obtained by simply combining in quadrature the parameter uncertainties.

<sup>17</sup>The LLNL energy dependence relationship, from Slide 84 of LLNL-PRES-419362, is:

$$Y_{147\text{Nd}}^{239\text{Pu},fs} = [(7.96 \pm 2.26) \times 10^{-4} \cdot \langle E_n \rangle] + (0.01955 \pm 0.00020)$$

$^{235}\text{U}$ ,  $^{238}\text{U}$ , and  $^{239}\text{Pu}$  in Table 14 of LA-UR 09-06679. Since the focus of the Panel is the latter target, Table 2 reproduces LANL's *experimental*  $Q_{99\text{Mo}}^{239\text{Pu},fs}$  along with *constructed* values using the  $Y_{99\text{Mo}}^{239\text{Pu},fs}$  from Table 1 above and  $Y_{99\text{Mo}}^{235\text{U},th}$  ( $6.108 \pm 0.086$ ) from ENDF/B-VI, which is a quantity that has been reported by many investigators and is considered to be well characterized.

Table 2: Comparison of *experimental* LANL  $Q_{99\text{Mo}}^{239\text{Pu},fs}$  with values *constructed* from  $Y_{99\text{Mo}}^{239\text{Pu},fs}$  and  $Y_{99\text{Mo}}^{235\text{U},th}$

LANL Experiment $\frac{K_{99\text{Mo}}^{235\text{U},th}}{K_{99\text{Mo}}^{239\text{Pu},fs}} = Q_{99\text{Mo}}^{239\text{Pu},fs}$	LANL Evaluation $\frac{Y_{99\text{Mo}}^{239\text{Pu},fs}}{Y_{99\text{Mo}}^{235\text{U},th}} = Q_{99\text{Mo}}^{239\text{Pu},fs}$	LLNL Evaluation $\frac{Y_{99\text{Mo}}^{239\text{Pu},fs}}{Y_{99\text{Mo}}^{235\text{U},th}} = Q_{99\text{Mo}}^{239\text{Pu},fs}$
$\frac{(2.445 \pm 0.039) \times 10^5}{(2.409 \pm 0.048) \times 10^5} = 1.015 \pm 0.026$	$\frac{6.23 \pm 0.06}{6.108 \pm 0.086} = 1.020 \pm 0.017$	$\frac{6.12 \pm 0.08}{6.108 \pm 0.086} = 1.002 \pm 0.019$

## B Accounting for $^{240}\text{Pu}$ content in LANL R values

The fission product yield  $Y_j^{i,e}$  is based on the measurement of two quantities:

1.  $F_j^{i,e}$ , the number of fissions induced in the sample during irradiation
2.  $N_j^{i,e}$ , the number of nuclei of fission product  $j$  produced in the sample

This Appendix describes how the presence of  $^{240}\text{Pu}$  in a Pu sample impacts these two observables, and how measured R values are similarly affected.

### 1 - Number of fissions

For the Pu samples under consideration, the overwhelming majority of the material is either  $^{239}\text{Pu}$  or  $^{240}\text{Pu}$ . The total number of fissions  $F^{\text{Pu},e}$  induced in the sample can be expressed as

$$F^{\text{Pu},e} = F^{^{239}\text{Pu},e} + F^{^{240}\text{Pu},e} \quad (1)$$

If we define  $N_{^{239}\text{Pu}}$  as the number of atoms of  $^{239}\text{Pu}$  in the sample, and similarly  $N_{^{240}\text{Pu}}$  as the number of atoms of  $^{240}\text{Pu}$ , the following equation describes  $F^{^{240}\text{Pu},e}$ :

$$F^{^{240}\text{Pu},e} = F^{^{239}\text{Pu},e} \cdot \frac{N_{^{240}\text{Pu}}}{N_{^{239}\text{Pu}}} \cdot \frac{\sigma_{(n,f)}^{^{240}\text{Pu},e}}{\sigma_{(n,f)}^{^{239}\text{Pu},e}} \quad (2)$$

where  $\sigma_{(n,f)}^{^{239}\text{Pu},e}$  and  $\sigma_{(n,f)}^{^{240}\text{Pu},e}$  are the fission cross sections for  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  in neutron field  $e$ .

Substituting (2) into (1), we obtain

$$F^{\text{Pu},e} = F^{^{239}\text{Pu},e} \left( 1 + \frac{N_{^{240}\text{Pu}}}{N_{^{239}\text{Pu}}} \cdot \frac{\sigma_{(n,f)}^{^{240}\text{Pu},e}}{\sigma_{(n,f)}^{^{239}\text{Pu},e}} \right) \quad (3)$$

### 2 - Number of fission products

Analogous to Equations (1) and (2), the following describe the total number of atoms of fission product  $j$  produced in the mixed  $^{239}\text{Pu}/^{240}\text{Pu}$  sample:

$$N_j^{\text{Pu},e} = N_j^{^{239}\text{Pu},e} + N_j^{^{240}\text{Pu},e} \quad (4)$$

$$N_j^{^{240}\text{Pu},e} = N_j^{^{239}\text{Pu},e} \cdot \frac{N_{^{240}\text{Pu}}}{N_{^{239}\text{Pu}}} \cdot \frac{\sigma_{(n,f)}^{^{240}\text{Pu},e}}{\sigma_{(n,f)}^{^{239}\text{Pu},e}} \cdot \frac{Y_j^{^{240}\text{Pu},e}}{Y_j^{^{239}\text{Pu},e}} \quad (5)$$

where  $Y_j^{^{239}\text{Pu},e}$  and  $Y_j^{^{240}\text{Pu},e}$  are the fission product yields for  $j$  in neutron field  $e$ . Substituting (5) into (4) produces

$$N_j^{\text{Pu},e} = N_j^{^{239}\text{Pu},e} \left( 1 + \frac{N_{^{240}\text{Pu}}}{N_{^{239}\text{Pu}}} \cdot \frac{\sigma_{(n,f)}^{^{240}\text{Pu},e}}{\sigma_{(n,f)}^{^{239}\text{Pu},e}} \cdot \frac{Y_j^{^{240}\text{Pu},e}}{Y_j^{^{239}\text{Pu},e}} \right) \quad (6)$$

### 3 - Fission product yield

Recalling the definition of the fission product yield,

$$Y_j^{i,e} = \frac{N_j^{i,e}}{F^{i,e}} \quad (7)$$

and substituting (6) and (3) into the numerator and denominator, respectively, we obtain the result describing the impact of  $^{240}\text{Pu}$  on  $Y_j^{\text{Pu},e}$ :

$$Y_j^{\text{Pu},e} = Y_j^{239\text{Pu},e} \frac{1 + \frac{N_{240\text{Pu}}}{N_{239\text{Pu}}} \cdot \frac{\sigma_{(n,f)}^{240\text{Pu},e}}{\sigma_{(n,f)}^{239\text{Pu},e}} \cdot \frac{Y_j^{240\text{Pu},e}}{Y_j^{239\text{Pu},e}}}{1 + \frac{N_{240\text{Pu}}}{N_{239\text{Pu}}} \cdot \frac{\sigma_{(n,f)}^{240\text{Pu},e}}{\sigma_{(n,f)}^{239\text{Pu},e}}} \quad (8)$$

As an example, consider the measurement of  $^{147}\text{Nd}$  in a sample containing 6 atom%  $^{240}\text{Pu}$ , irradiated in the Flattop-25 critical assembly. In this case,

$$\frac{N_{240\text{Pu}}}{N_{239\text{Pu}}} \approx 0.064$$

$$\frac{Y_{147\text{Nd}}^{240\text{Pu,Flattop-25}}}{Y_{147\text{Nd}}^{239\text{Pu,Flattop-25}}} \approx 1.1 \quad (\text{from Myers}^9)$$

$$\frac{\sigma_{(n,f)}^{240\text{Pu,Flattop-25}}}{\sigma_{(n,f)}^{239\text{Pu,Flattop-25}}} \approx 0.55^{18}$$

Substituting these values into (8) gives

$$Y_{147\text{Nd}}^{\text{Pu,Flattop-25}} = Y_{147\text{Nd}}^{239\text{Pu,Flattop-25}} \times 1.0034$$

Thus, the measured  $^{147}\text{Nd}$  fission product yield in the sample would exceed that of pure  $^{239}\text{Pu}$  by  $\approx 0.35\%$ .

### 4 - Impact on R values

Recall that an R value can be expressed in terms of fission product yields,

$$R_j^{i,e} = \frac{Q_j^{i,e}}{Q_{99\text{Mo}}^{i,e}} = \frac{\frac{Y_j^{i,e}}{Y_j^{235\text{U},th}}}{\frac{Y_{99\text{Mo}}^{i,e}}{Y_{99\text{Mo}}^{235\text{U},th}}}$$

If we accept the widely held belief that the fission product yield of  $^{99}\text{Mo}$  is essentially the same for  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  in a fission spectrum neutron field, the impact of  $^{240}\text{Pu}$  on measured R values will be comparable to the impact on the fission product yield. Using the example above,

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<sup>18</sup>LANL experimental value = 0.549, MCNP calculation = 0.573

the  $^{147}\text{Nd}$  R value for a sample containing  $\approx 6$  atom%  $^{240}\text{Pu}$ , and irradiated in the Flattop-25 critical assembly, would also be  $\approx 0.35\%$  higher than for pure  $^{239}\text{Pu}$ .

Finally, it should be recognized that because of the energy dependent relationship of  $\frac{\sigma_{(n,f)}^{240\text{Pu,e}}}{\sigma_{(n,f)}^{239\text{Pu,e}}}$  (see Figure 1), the impact of  $\approx 6$  atom%  $^{240}\text{Pu}$  in a sample irradiated by a “harder” neutron spectrum (such as Jezebel) would be on the order of  $0.6\%$ .

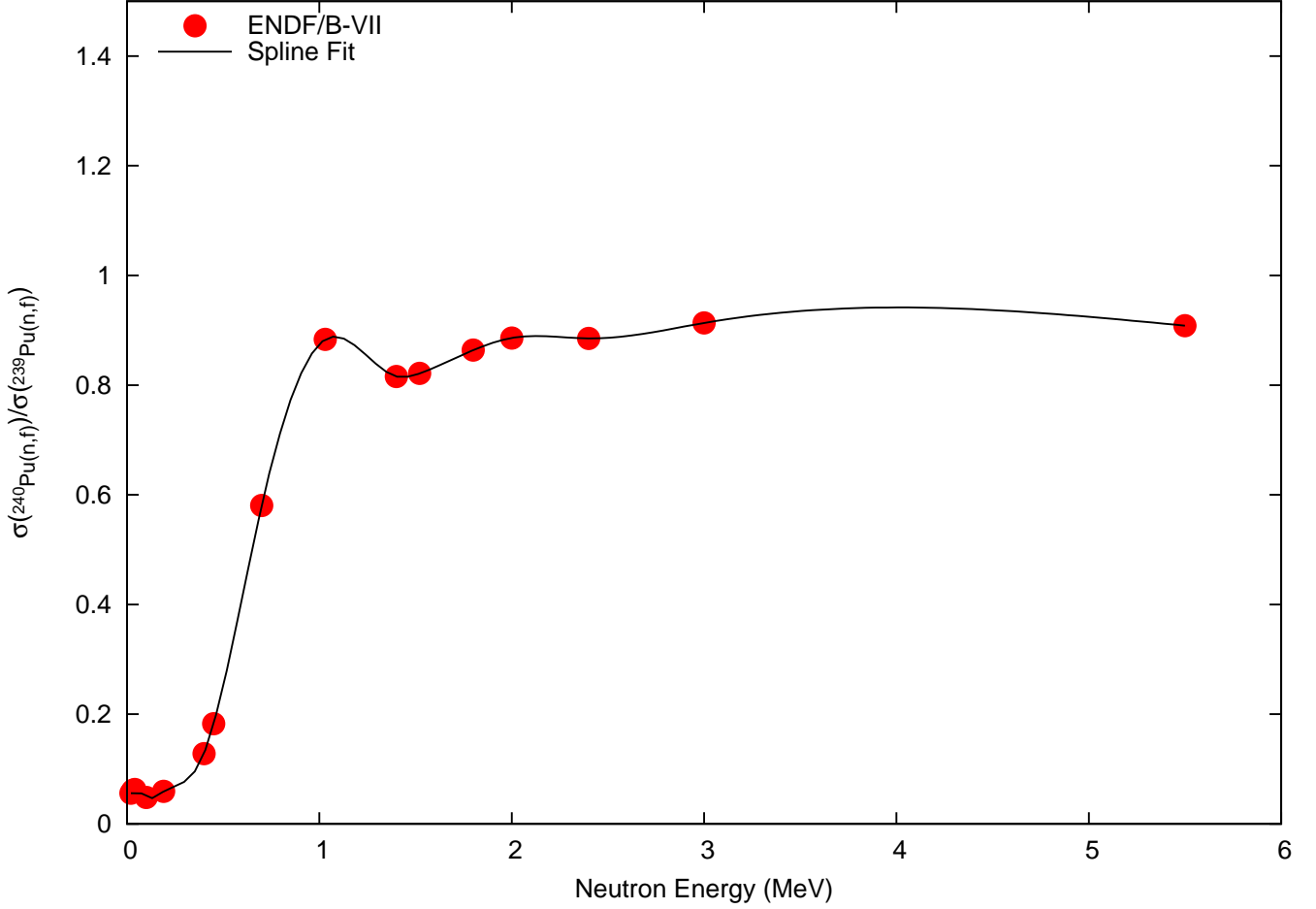


Figure 1: Relationship between fission rates for  $^{240}\text{Pu}$  and  $^{239}\text{Pu}$  as a function of energy.

# Glossary

**Fission Product (FP):** A nuclide, often radioactive, produced as the result of a nuclear fission event.

**Fission Product Yield:** The ratio of the number of atoms ( $N_j^{i,e}$ ) of a particular fission product nuclide  $j$  produced per fission event ( $F^{i,e}$ ) of target  $i$  in neutron field  $e$

$$Y_j^{i,e} = \frac{N_j^{i,e}}{F^{i,e}}$$

The units of  $Y_j^{i,e}$  are atoms/fission.

**Fission Spectrum:** In the context of this report, a “fission spectrum” is a source of neutrons where the average/most probable neutron energy may range from a few keV to a few MeV. The spread in neutron energies about the average/most probable energy in any given source, while largely consistent with semi-empirical functional forms (like the Watt function), may show deviations from such simple relationships. Since detailed experimental measurements of “fission spectra” are scarce, synthetic spectra (e.g. from MCNP calculations) and integral “spectral indices” must be used to estimate differences between various “fission spectrum” sources.

**K Factors:** These relate, in a given sample, total fissions produced by irradiation of target  $i$  in neutron field  $e$  to the induced activity of fission product  $j$ .

$$K_j^{i,e} = \frac{F^{i,e}}{A_j^{i,e}}$$

where

$F^{i,e}$  is the total number of “fissions” in the sample under study

$A_j^{i,e}$  is the observed activity, reported in units of counts per minute (cpm), for fission product  $j$ , on a particular detector.  $A_j$  is corrected for decay occurring both during bombardment and between end of bombardment and measurement, as well as for losses during chemical processing.

The units of  $K_j^{i,e}$  are fissions/cpm.

**Neutron field:** In this report, the expression “neutron field” is used to describe the type of irradiation environment to which a fissionable target is exposed. In particular, “fission spectrum” (abbreviated as *fs*) and “thermal” (abbreviated as *th*) neutron fields are referenced. Other Glossary entries provide working definitions of these two neutron fields.

**Q Values:** These relate the K factor for any fissionable isotope,  $i$ , and neutron field,  $e$ , to the K for  $^{235}\text{U}$  in a thermal neutron field, for the same FP isotope,  $j$ .

$$Q_j^{i,e} = \frac{K_j^{235\text{U},th}}{K_j^{i,e}}$$

It should be noted that  $Q_j^{i,e}$  may also be defined in terms of fission product yields,

$$Q_j^{i,e} = \frac{Y_j^{i,e}}{Y_j^{235\text{U},th}}$$

$Q_j^{i,e}$  values are dimensionless.

**r Values (lower case):** Activity ratios of a given fission product nuclide  $j$  to a reference fission product nuclide (often  $^{99}\text{Mo}$ ), resulting from the bombardment of fissionable target  $i$  in neutron field  $e$ .

$$r_j^{i,e} = \frac{A_j^{i,e}}{A_{99\text{Mo}}^{i,e}}$$

$r_j^{i,e}$  values are dimensionless.

**R Values (upper case):** These relate ratios of activities of different fission product nuclides from bombardment of a common fissioning isotope in a common neutron field to the ratio of these same fission products from thermal neutron fission of  $^{235}\text{U}$ . The reference fission product is often  $^{99}\text{Mo}$ , but is not required to be so.

$$R_j^{i,e} = \frac{\frac{A_j^{i,e}}{A_{99\text{Mo}}^{i,e}}}{\frac{A_j^{235\text{U},th}}{A_{99\text{Mo}}^{235\text{U},th}}} = \frac{r_j^{i,e}}{r_j^{235\text{U},th}}$$

Note that  $R_j^{i,e}$  may be defined in terms of  $Q_j^{i,e}$ ,

$$R_j^{i,e} = \frac{Q_j^{i,e}}{Q_{99\text{Mo}}^{i,e}}$$

$R_j^{i,e}$  values are dimensionless.

**Thermal neutrons:** A neutron field whose energy spectrum may be approximated by a Maxwell-Boltzmann distribution with a “most probable energy” of 0.025 eV.